

09/914 994

COPC

Patent No. 6,913,738
Request for Cert. of Correction dated January 11, 2006
Attorney Docket No. 0388-011500



IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Patent No. : 6,913,738 Confirmation No. 7881
Inventor : Echigo et al.
Issued : July 5, 2005
Title : System For Removing Carbon Monoxide and
Method For Removing Carbon Monoxide
Examiner : Ngoc-Yen Nguyen
Customer No. : 28289

REQUEST FOR CERTIFICATE OF CORRECTION OF PATENT
FOR PTO MISTAKE (37 C.F.R. 1.322(a))

Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

ATTENTION: Decision and Certificate of Correction Branch
Patent Issue Division

Sir:

In accordance with 35 U.S.C. §254, we attach hereto Form PTO/SB/44 and a copy of proof of PTO errors and request that a Certificate of Correction be issued in the above-identified patent. The following errors appear in the patent as printed:

Face of the Patent, See Item (57) ABSTRACT, line 9, "are co-existent For" should read -- are co-existent. For --
(The period is missing after "co-existent". See application, page 28, line 9.)

Column 3, line 5, "methanation rein oval" should read -- methanation removal --
(See application, page 5, line 9.)

Column 6, line 28, "dower than 240°C" should read -- (lower than 240°C) --
(See application, page 11, line 7.)

Column 7, DELETE THE TEXT FROM: line 5, beginning with "In each catalyst" and ending at line 42 "supported on an alumina".
(This text was duplicated from the original specification page 12 which was replaced with an amended page 12.)

Column 8, line 36, "dower than 250°C" should read -- (lower than 250°C) --
(See application page 13, line 7.)

Column 8, line 36, "6chemical" should read -- 6 chemical --.
(See application, page 14, lines 1-2.)

Column 8, line 52, "H₂O, etc. carbon" should read -- H₂O, etc. So that carbon --
(See application, page 14, lines 14-15.)

*Certificate
JAN 20 2006
of Correction.*

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Column 10, line 18, "as the value "0"" should read -- as for the value "0" --
(See application, page 17, lines 5-6.)

Column 10, line 23, "the first CO remover the" should read
-- the first CO remover for the --
(See application, page 17, lines 8-9.)

Column 10, line 40, "first CO remover" should read -- first CO remover 6. --
(See application, page 17, lines 14-15.)

Column 10, line 48, "humidified gas" should read -- (humidified gas --
(See application, page 18, line 5.)

Column 12, line 49, "of 135C" should read -- of 135°C --
(See application, page 21, line 30.)

Column 13, line 21, "remover 6 composing" should read -- remover 6 comprising --
(See application, page 23, line 3.)

Respectfully submitted,

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UNITED STATES PATENT AND TRADEMARK OFFICE
CERTIFICATE OF CORRECTION

Page 1 of 2

PATENT NO. : 6,913,738 B1
 APPLICATION NO. : 09/914,994
 ISSUE DATE : July 5, 2005
 INVENTORS : Echigo et al.

It is certified that an error appears or errors appear in the above-identified patent and that said Letters Patent is hereby corrected as shown below:

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MAILING ADDRESS OF SENDER: The Webb Law Firm
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This collection of information is required by 37 CFR 1.322, 1.323, and 1.324. The information is required to obtain or retain a benefit by the public which is to file (and by the USPTO to process) an application. Confidentiality is governed by 35 U.S.C. 122 and 37 CFR 1.14. This collection is estimated to take 1.0 hour to complete, including gathering, preparing, and submitting the completed application form to the USPTO. Time will vary depending upon the individual case. Any comments on the amount of time you require to complete this form and/or suggestions for reducing this burden, should be sent to the Chief Information Officer, U.S. Patent and Trademark Office, U.S. Department of Commerce, P.O. Box 1450, Alexandria, VA 22313-2450. DO NOT SEND FEES OR COMPLETED FORMS TO THIS ADDRESS. SEND TO: Attention Certificate of Corrections Branch, Commissioner for Patents, P.O. Box 1450, Alexandria, VA 22313-1450.

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UNITED STATES PATENT AND TRADEMARK OFFICE
CERTIFICATE OF CORRECTION

Page 2 of 2

PATENT NO. : 6,913,738 *B1*
APPLICATION NO. : 09/914,994
ISSUE DATE : July 5, 2005
INVENTORS : Echigo et al.

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See "PA"

SPECIFICATION

SYSTEM AND METHOD FOR REMOVING CARBON MONOXIDE

See "PA"

See "PA"

5 1. Field of the Invention

TECHNICAL FIELD

The present invention relates to the technique of removing carbon monoxide contained in a hydrogen-rich reformed gas (an example of "treatment-object gas" as so referred to in the present application) such as 10 obtained e.g. in the reforming process of hydrocarbon fuels including natural gas, naphtha, kerosene, etc, or alcoholic fuels such as methanol.

The technique to which the present application relates is characterized that it can remove carbon monoxide up to a concentration of ten ppm or lower. For this ability, the technique can be suitably employed 15 in a power generating system using e.g. solid polymer electrolyte fuel cell which operates at a relatively low temperature.

For the purpose of simplifying the description, the following description will be made by taking a reformed gas used in a fuel cell as an example of the treatment-object gas.

20 See "PA"

2. Description of Related Art

BACKGROUND ART

Conventionally, with a fuel reforming apparatus using fossil fuel such as natural gas as raw fuel, a carbon monoxide shift converter is 25 connected to the downstream end of the reformer so as to convert carbon monoxide in the reformed gas into carbon dioxide by the water-gas shift reaction, whereby the carbon monoxide concentration is reduced (removed) to 1% approximately.

On the other hand, with a fuel reforming apparatus using methanol 30 as raw fuel, since this apparatus involves a step of the water-gas shift

can be a useful fuel will be lost by combustion.

(b) problem with removal using methanator

5 With this technique, if the treatment-object gas contains also carbon dioxide as is the case with a reformed gas, methanation of carbon dioxide, in addition to that of carbon monoxide, tends to occur with very high likelihood. For this reason, if carbon monoxide is to be removed sufficiently while restricting loss of hydrogen due to methanation of carbon dioxide, it is
10 necessary to first absorb and remove the carbon dioxide also present in the reformed gas, so that the system required for this tends to be complicated.

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SUMMARY OF THE INVENTION
DISCLOSURE OF THE INVENTION

15 The present invention has been made in order to solve the above-described problems, and its object is to obtain a carbon-monoxide removing technique capable of very effectively reducing/removing carbon monoxide present at one thousand of ppm to several % in a hydrogen-rich treatment-object gas such as a reformed gas obtained by reforming of a fuel such as
20 natural gas, methanol, etc. to a concentration of several tens of ppm (preferably 10 ppm) or lower without excessive loss of hydrogen (with minimizing the consumption of hydrogen), even when carbon dioxide, methane are co-existent.

25 For accomplishing this object, according to characterizing features of the present invention, a system for removing carbon monoxide from a hydrogen-containing treatment-object gas comprises two stages of CO removers for removing carbon monoxide, the first-stage CO remover removing a portion of the carbon monoxide by methanation thereof through a catalyst reaction, the second-stage CO remover removing the remaining
30 portion of the carbon monoxide mainly by oxidation thereof through a

further catalyst reaction involving addition of an oxidizing agent.

The carbon monoxide removing system of the invention includes two stages of first CO remover and second CO remover which are disposed in the mentioned order, so that the treatment-object gas containing carbon 5 monoxide is fed first into the first CO remover and then into the second CO remover, whereby treatment-object gas having its carbon monoxide content removed is obtained from the second CO remover.

In the above, the removal of carbon monoxide by the first CO 10 remover is methanation removal using catalyst reaction and that by the second CO remover is mainly oxidation removal using catalyst reaction involving addition of an oxidizing agent.

Accordingly, in this removing process, at the first CO remover, by 15 using hydrogen present in the surrounding, methanation of carbon monoxide is promoted for removal of the carbon monoxide, so that no oxidizing agent is required. By this catalyst reaction, a major part (more than half) of carbon monoxide present in the treatment-object gas may be methanated to be removed.

Subsequently, at the second CO remover, the remaining portion of 20 the carbon monoxide is removed mainly through oxidation thereof by a catalyst reaction involving addition of an oxidizing agent. In this case, since the amount of the carbon monoxide has already been reduced, the remaining amount of carbon monoxide can be substantially entirely removed (to a concentration of several ppm approximately, for instance) with restricting the amount of the oxidizing agent to be added to the treatment-object gas.

25 Therefore, with this carbon-monoxide removing system, it is possible to restrict the amount of the oxidizing agent required for the removal to be smaller than the equivalent of the carbon monoxide entering the first CO remover. As a result, treatment-object gas free from carbon monoxide may be obtained with limiting the amount of useful hydrogen to be 30 consumed in the combustion.

monoxide as methane. In doing this, it is preferred that the methanation reaction be effected with setting the methanation reaction temperature higher than 160°C and lower than 240°C.

With this, by setting the temperature higher than a predetermined temperature (higher than 160°C), the methanation reaction will proceed to a certain degree, whereas by setting also this temperature lower than a predetermined temperature (lower than 240°C), it is possible to sufficiently restrict occurrence of methanation of carbon dioxide which tends to involve consumption of hydrogen. More preferably, the upper-limit temperature is set at 200°C.

In this case, such relatively low temperature range is employed for the methanation reaction. Therefore, it is preferred from the view point of catalyst reactivity, the first metal catalyst comprise catalyst containing Ru.

15 BRIEF DESCRIPTION OF THE DRAWING

Fig. 1 is a view showing a first embodiment.

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DETAILED DESCRIPTION OF THE PREFERRED EMBODIMENTS BEST MODE OF EMBODYING THE INVENTION

20

An example of mode of using a carbon-monoxide removing system according to the present invention will be described.

From a carbon-monoxide shift converter reactor, a treatment-object gas which contains a relatively large amount, i.e. about 6000 ppm to 1 wt.%, of carbon monoxide is guided to a first CO remover (incorporating a "high quantity metal supported catalyst"). Generally, this treatment-object gas contains no oxidizing component (oxygen).

Next, the treated treatment-object gas discharged from the first CO remover is guided to a second CO remover. In this second CO remover (incorporating a "low quantity metal supported catalyst"), to a trace amount

(incorporating a "low quantity metal supported catalyst"), to a trace amount of carbon monoxide which remains un-removed at the first CO remover, air or oxygen alone is added as an oxidizing agent to obtain $[O_2] / [CO]$ ratio of 0.5 to 4.5 and then a reaction is carried out at a relatively low temperature range. In this, a CO sensor or the like may be provided between the second CO remover and the first CO remover so that the amount of oxidizing agent be controlled based on a detection value from this sensor.

The "high quantity metal supported catalyst" is a catalyst comprising 0.5 to 5 wt.% of one or more kinds of metal selected from the group consisting of Ru, Pt, Rh, Pd and Ni supported on an alumina. The "low quantity metal supported catalyst" is a catalyst comprising 0.1 to 2 wt.% of one or more kinds of metal selected from the group consisting of Ru, Pt, Rh and Pd supported on an alumina.

In each catalyst reaction, a value of GHSV (Gas Hourly Space Velocity: treatment-object gas flow amount/ catalyst volume (1/h)) is set to about 500 to 100000/h (set to a practically possible range).

Further, the reaction temperature (°C) at the first CO remover is set to a range from 155 to 300°C. Whereas, the reaction temperature at the second CO remover is set to a range from 50 to 250°C lower than the reaction temperature at the first CO remover. That is, the former is set to be higher than the latter.

In the above, preferably, the reaction temperature of the first step at the first CO remover is set to 155 to 300°C (more preferably, to 175 to 250°C). And, preferably, the reaction temperature of the second step at the second CO remover is set to a relatively lower range of 50 to 250°C (more preferably, to 100 to 160°C). This is because the temperature range should differ in correspondence with each object.

If the temperature of the first step is lower than 155°C, the methanation activity tends to be lower. Whereas, if it is higher than 300°C, an influence of a side reaction tends to appear. Then, if the reaction

of carbon monoxide which remains un-removed at the first CO remover, air or oxygen alone is added as an oxidizing agent to obtain $[O_2] / [CO]$ ratio of 0.5 to 4.5 and then a reaction is carried out at a relatively low temperature range. In this, a CO sensor or the like may be provided between the second 5 CO remover and the first CO remover so that the amount of oxidizing agent be controlled based on a detection value from this sensor.

The "high quantity metal supported catalyst" is a catalyst comprising 0.5 to 5 wt.% of one or more kinds of metal selected from the group consisting of Ru, Pt, Rh, and Pd supported on an alumina. The "low 10 quantity metal supported catalyst" is a catalyst comprising 0.1 to 2 wt.% of one or more kinds of metal selected from the group consisting of Ru, Pt, Rh and Pd supported on an alumina support.

In each catalyst reaction, a value of GHSV (Gas Hourly Space Velocity: treatment-object gas flow amount/ catalyst volume (1/h)) is set to 15 about 500 to 100000/h (set to a practically possible range).

Further, the reaction temperature ($^{\circ}C$) at the first CO remover is set to a range from 155 to 300 $^{\circ}C$. Whereas, the reaction temperature at the second CO remover is set to a range from 50 to 250 $^{\circ}C$ lower than the reaction 20 temperature at the first CO remover. That is, the former is set to be higher than the latter.

In the above, preferably, the reaction temperature of the first step at the first CO remover is set to 155 to 300 $^{\circ}C$ (more preferably, to 175 to 250 $^{\circ}C$). And, preferably, the reaction temperature of the second step at the second CO remover is set to a relatively lower range of 50 to 250 $^{\circ}C$ (more 25 preferably, to 100 to 160 $^{\circ}C$). This is because the temperature range should differ in correspondence with each object.

If the temperature of the first step is lower than 155 $^{\circ}C$, the methanation activity tends to be lower. Whereas, if it is higher than 300 $^{\circ}C$, an influence of a side reaction tends to appear. Then, if the reaction

temperature is set to be lower than 250°C, methanation of carbon dioxide which is unnecessary in the present invention, can be restricted in particular. For achieving the restriction of methanation of carbon dioxide and promotion of methanation of carbon monoxide, an even more preferred range is a 5 temperature range from 160°C to 240°C.

On the other hand, at the second step, if the temperature range is set to be relatively low (lower than 250°C), oxidation reaction will mainly take place, so that it becomes easier to reduce carbon monoxide to a sufficient level.

10 If the temperature of the second step is lower than 50°C, the reactivity will be low. Whereas, if it is higher than 250°C, it may happen that it becomes difficult to reduce carbon monoxide to be lower than several tens of ppm, due to a side effect such as a reverse-shift reaction (reverse water-gas shift reaction).

15 With the above arrangements, at the first CO remover, carbon monoxide is reacted with hydrogen in the treatment-object gas to be converted into methane at the relatively high temperature range according to a reaction formula: $\text{CO} + 3\text{H}_2 \rightarrow \text{CH}_4 + \text{H}_2\text{O}$, so that most of the carbon monoxide may be removed. This reaction can take place through 20 appropriate control of the catalyst reaction temperature, substantially without aid of an oxidizing agent. In this case, the amount of carbon monoxide removable by the methanation reaction can be higher than 70% of that introduced into the first CO remover.

Next, at the second CO remover, mainly through a oxidation 25 reaction involving an oxidizing agent according to a formula: $2\text{CO} + \text{O}_2 \rightarrow 2\text{CO}_2$, carbon monoxide is removed. This removal is possible to a level of several tens of ppm (preferably, 10 ppm) or lower. Hence, this may be suitably applied to a polymer electrolyte fuel cell.

Accordingly, for carbon monoxide contained in a treatment-object 30 gas at a reaction outlet of a carbon monoxide shift converter, the

conventional method requires oxygen three times in the mole ratio, i.e. 6 chemical equivalents. On the other hand, according to the present invention, only with addition of air containing oxygen by a concentration lower than the chemical equivalent of the carbon monoxide, the carbon monoxide contained in the treatment-object gas may be removed. And, unnecessary consumption of hydrogen may be reduced correspondingly.

Further, even when air is selected as the oxidizing agent, the addition amount of air is small. Thus, the amount of nitrogen to be mixed into the treatment-object gas too can be reduced. Consequently, reduction in the partial pressure of the hydrogen in the treatment-object gas may be decreased.

By appropriate control of the reaction temperatures of the first CO remover and the second CO remover, it is possible to restrict such side reaction as: $\text{CO}_2 + 4\text{H}_2 \rightarrow \text{CH}_4 + 2\text{H}_2\text{O}$ or $\text{CO}_2 + \text{H}_2 \rightarrow \text{CO} + \text{H}_2\text{O}$, etc. So that, carbon monoxide may be removed very efficiently even when several tens of % of carbon dioxide is co-existent in the gas and the loss of hydrogen may be reduced.

The removing method of the invention is very suitable also for a case where methane is present in the treatment-object gas, since the method functions well in such case as well.

Further, comparing the reaction temperature of the first CO remover and the reaction temperature of the second CO remover to each other, the reaction temperature shifts from a high temperature to a low temperature along the flow passage.

Moreover, if the treatment-object gas is supplied to a low-temperature operating fuel cell such as a solid polymer electrolyte fuel cell by employing the method of the present invention, it is possible to supply fuel gas with lower efficiency reduction, by avoiding CO poisoning of the electrode catalyst of the fuel cell.

Table 1

example	first CO remover temperature (°C)	SV value 1/h	exit CO concentration (ppm)	exit CO concentration of second CO remover (ppm)
1	200	3750	353	0
2	200	5000	1627	0
3	210	5000	322	0
4	210	7500	1497	0
5	230	7500	223	0
6	230	15000	889	0

5 In the above, the CO concentration of the treatment-object gas introduced into the first CO remover was 6000 ppm. In the table above, as for the value "0" as the CO concentration, the detection limit of CO concentration was 5 ppm.

Now, the concentrations of methane formed at the first CO remover for the respective examples were as follows.

10

example	first CO remover temperature (°C)	exit CO concentration (ppm)	concentration of methane formed (ppm)
1	200	353	7013
2	200	1627	4483
3	210	322	7179
4	210	1497	4577
5	230	223	10145
6	230	889	10803

The results show that CO removal was possible in each case with an amount of oxygen (the amount of oxidizing agent) smaller than the chemical equivalent of the amount of carbon monoxide entering the first CO remover

15

6.

[second embodiment]

The first CO remover 6 was charged with catalyst (as a first metal catalyst and also as a high quantity metal supported catalyst) comprising 2 wt.% of ruthenium supported on granular alumina. Then, a reformed gas (humidified gas containing 6000 ppm of carbon monoxide, 5000 ppm of methane, 20% of carbon dioxide and 78.9% of hydrogen) obtained from the exit of carbon-monoxide shift converter 5 was introduced to this first CO remover 6, in which a methanation reaction of CO was effected at GHSV 3750-5000/h and at a temperature of 220 to 260°C.

Next, the second CO remover 7 was charged with catalyst (as a second metal catalyst and also as a low quantity metal supported catalyst) comprising 1 wt.% of ruthenium supported on granular alumina. Then, the reformed gas from the exit of the first CO remover 6 was introduced to this second CO remover 7, in which with addition of air containing oxygen by an amount corresponding to $[O_2] / [CO]$ ratio of 1.3 relative to the CO concentration of the reformed gas at the entrance to this second CO remover 7, CO oxidation reaction was carried out at GHSV 15000/h approximately and at a temperature of 135°C.

The results are summarized and shown in Table 2 below.

20

Table 2

example	first CO remover temperature (°C)	SV value 1/h	exit CO concentration (ppm)	exit CO concentration of second CO remover (ppm)
1	220	3750	1021	0
2	220	5000	1510	0
3	240	3750	803	0
4	240	5000	965	0
5	260	5000	1053	0

CO remover are provided separately from each other. Instead, it is possible to provide, as a construction for effecting the above-described process, a single-container construction housing a catalyst for methanation disposed on the upstream side in the flow direction of the treatment-object gas having catalyst for methanation, a catalyst for oxidation disposed on the downstream side in the same direction with a mechanism for introducing an oxidizing agent to this portion.

In such case, the upstream portion of the container corresponds to the first CO remover and the downstream portion thereof corresponds to the second CO remover.

(d) Other embodiments of the present invention will be described next.

[other embodiments]

15

The first CO remover 6 was charged with catalyst (as a first metal catalyst) comprising a granular alumina supporting 1 wt.% of rhodium. Then, a reformed gas (same as first and second embodiments) obtained from the exit of carbon-monoxide shift converter 5 was introduced to this first CO remover 6, in which a methanation reaction of CO was effected at GHSV 3750-7500/h and at a temperature of 260 to 300°C.

Next, the second CO remover 7 was charged with catalyst (as a second metal catalyst and also as a low quantity metal supported catalyst) comprising 1 wt.% of ruthenium supported on granular alumina. Then, the reformed gas from the exit of the first CO remover 6 was introduced to this second CO remover 7, in which with addition of air 8 containing oxygen by an amount corresponding to $[O_2] / [CO]$ ratio of 1.3 relative to the CO concentration of the reformed gas at the entrance to this second CO remover 7, CO oxidation reaction was carried out at GHSV 15000/h approximately and at a temperature of 135°C.

can be employed in the second CO remover in this invention.

Further, by using the combination shown in the first alternate embodiment of the first CO remover 6 comprising the rhodium catalyst and the second CO remover 7 comprising the ruthenium catalyst and under the 5 conditions of example 5 (the example shown as example 5 in Table 3), the catalyst of the second CO remover 7 was replaced by 1 wt.% of rhodium supported on granular alumina (an example of second metal catalyst). Then, the system was operated.

The operating conditions of the second CO remover 7 were: the 10 temperature 250°C; GHSV 15000 and the addition amount of air: $[O_2] / [CO] = 4$. With these, in this case too, the carbon monoxide concentration at the exit of the second CO remover 7 was reduced to 0 ppm (below the actual detection limit). Therefore, rhodium can be employed in the second CO remover in this invention.

15

to "PA"

~~EFFECT OF THE INVENTION~~

According to the present invention, the amount of oxidizing agent such as air or oxygen to be added in the course of removal of carbon 20 monoxide from a reformed gas can be reduced significantly. Thus, CO removal of reformed gas is possible with minimizing loss of hydrogen by combustion.

Further, since in a fuel cell system methane produced by methanation of carbon monoxide can be used as a fuel for a burner of the 25 reformer, it is possible to improve high efficiency of the system.

For this reason, it is possible to feed a fuel reformed gas with high efficiency to a low-temperature operating type fuel cell such as polymer electrolyte fuel cell using a fuel such as natural gas, methanol, etc.

Since the above-described method of the present invention allows 30 efficient removal of carbon monoxide with a relatively high GHSV, the

ABSTRACT OF THE DISCLOSURE

The object of the present invention is to obtain a carbon-monoxide removing technique capable of very effectively reducing/removing carbon monoxide present at one thousand of ppm to several % in a hydrogen-rich treatment-object gas such as a reformed gas obtained by reforming of a fuel such as natural gas, methanol, etc. to a concentration of several tens of ppm (preferably 10 ppm) or lower without excessive loss of hydrogen, even when carbon dioxide, methane are co-existent. For accomplishing this object, 5 there are provided two stages of CO removers for removing carbon monoxide from a hydrogen-containing treatment-object gas, the first-stage CO remover removing a portion of the carbon monoxide by methanation thereof through a catalyst reaction, the second-stage CO remover removing the remaining portion of the carbon monoxide mainly by oxidation thereof through a 10 further catalyst reaction involving addition of an oxidizing agent. 15